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(54) Improvements of processes and plants for the production of urea.

(57) To the conventional process for the production of urea, comprising a stage for the reaction of NH_3 and CO_2 , a stage for stripping with CO_2 the urea solution containing carbamate and other products, a stage for condensing the stripped vapours and two stages of vacuum distillation, is now added at least a further stage selected from the group consisting of: a mean pressure distillation stage with substantially recovery heat, more particularly with double effect, a pre-distillation stage and a second stripping stage.

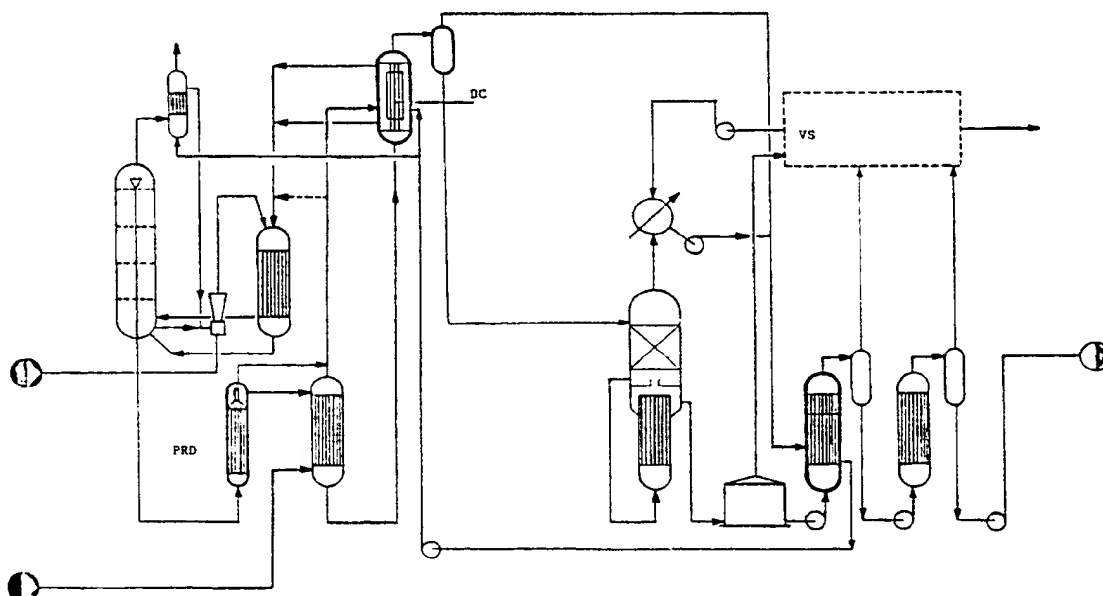


Fig. 7

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IMPROVEMENTS OF PROCESSES AND PLANTS FOR THE PRODUCTION OF UREA

Background of the invention1. Field of the invention

This invention relates to improvements to processes and plants for the production of urea, whereby ammonia and carbon dioxide are converted and the urea solution leaving the reactor is stripped with feed CO₂.

2. Description of Prior Art

Processes and plants of this type are widely used and have been remarkably successful, one reason being that besides having a single stripping stage with CO₂, they also have:

- a relatively low synthesis pressure (for example, about 140 bar);
- a high degree of depuration of the urea solution directly in the stripper which generally operates at the same synthesis pressure;
- a limited number of downstream stages for finishing the solution (for example stage at 3.5-4 bar, flash at 1 bar, vacuum and prilling sections).

Even with these undeniable advantages, the systems in question are not without drawbacks, particularly in respect of the consumption of process vapour which is certainly not low (for example 900 kg/t), and which, owing to the increase in the cost of energy due for example to the oil crisis, has become so significant as to constitute a handicap where continuing operation of existing plants is concerned. Numerous (and sometimes brilliant) attempts have been made to improve process efficiency but in almost all cases no concrete results have been achieved to date in the sense that more particularly energy consumption has not been reduced by any appreciable amount.

Moreover, owing to the low NH₃/CO₂ ratio in the reactor, conversion yields in the outlet reactor are low, thus making further developments more difficult.

The purpose of this invention is to improve the above systems for the production of urea, so as to achieve a drastic reduction in vapour consumption and more particularly to permit a simple and easy modernization of existing plants.

This and other aims are now surprisingly achieved by the addition of at least one stage selected from the group comprising: a mean pressure distillation stage using substantially recovery heat, preferably with double effect, and a pre-distillation stage upstream of the existing stripper.

The characteristics of the various embodiments are described in the claims, while the various aspects and advantages of the invention will be better illustrated by the description of the figures and by the attached tables.

PRIOR ART: STRIPPING PROCESS WITH THE AID OF CO₂

Figure 1 shows the simplified scheme of the conventional process. R indicates the reactor fed with NH₃ from line 1, through the ejector, condenser C1 and line 10, and with CO₂ from line 2, through stripper ST, condenser C1 and line 9.

The urea solution leaving reactor R flows by gravity (line 3) into stripper ST where the majority of carbamate and of free NH₃ are stripped by the feed CO₂ (line 2) and recycled to the reactor (line 4), after partial condensation in the film carbamate condenser C1, producing vapour at, for example, 4.5 bar abs.

From condenser C1, the recycle carbamate solution (8) plus a portion (30) of solution taken from the bottom of the reactor itself are sent to reactor R through ejector E.

The driving agent is the feed NH₃ (1). The residual vapours leaving from the top 7 of reactor R are sent to the scrubber HP (V.C.S.), where they are scrubbed with the return carbamate solution (20) and subsequently pass into a recovery section known per se and not described.

The solution leaving stripper ST is expanded directly in stage L.P., operating, for example, at about 3.5

bar, where it is distilled in D1 with vapour at, for example, 4.5 bar. Subsequently, it is expanded into the flash tank SB at 1 bar and from here goes into the vacuum distillers D2 and D3. Each distiller D1, D2, D3 is coupled to a separator S1, S2, respectively S3. The urea solution leaving S3 is sent to a crystallization section (known per se), while the vapours are condensed in a vacuum section VS to which also flow the vapours from tank SB.

Conventionally, the formation water is removed from 21 while the solution containing NH₃ and CO₂ is recycled to stage LF through 19.

For both stripper ST and the second vacuum distiller D3 the vapour used is at 20 bar, whereas recovery vapour at 4.5 bar is used for the other apparatus.

The NH₃ recovery section and the vacuum section are of the conventional type and they are not described here.

The typical operating conditions of the process are:

- Mol. ratio NH₃/CO₂ in the reactor : 2.8
- Mol. ratio H₂O/CO₂ in the reactor : 0.4
- Reactor yield : 57%
- Temperature at reactor outlet : 183–185 °C
- Reactor pressure : 141 bar
- Steam consumption at 20 bar : 1000 kg/t
- Export recovery steam at 4.5 bar : 350 kg/t.

The data relating to the conventional process are shown in Table 1.

SUMMARY OF THE INVENTION

Modernization of the conventional process

It has been found that a first improvement in energy terms in process efficiency can be obtained by adding a mean pressure (= 20 bar) stage in which the distillation heat is provided by the recovery heat/steam, while the vapours obtained are condensed in the first vacuum distiller, thus supplying an important part of the heat necessary for distillation.

Distillation heat can be obtained either by directly condensing the carbamate or with steam at 6 bar at least.

In the first type of embodiment (condensation of vapours to form carbamate, Fig. 2) a distiller-condenser DC is introduced, preferably of the type depicted in Fig. 3.

As Figure 2 shows, DC receives a part of the vapours from stripper ST (line 31) and the carbamate solution CA from the bottom of D2 through line 32. The condensate from DC goes to carbamate condenser C1 through line 34, while the urea solution from the bottom of ST is sent to be distilled through 35 in DC, followed by separator S4, whose vapours in 36 are sent to condense in D2 according to the double effect technique, known per se. In effect, the vapours from 36 (DC) are condensed in the presence of a weak carbamate solution 37 in D2 and the heat thus released is used to distill the urea solution flowing in a vacuum inside the D2 tubes.

As Figure 3 shows, it is a condenser-distiller DC in which condensation of the vapours from 31 takes place on the shell side M and the resulting heat is used to distill the urea solution flowing inside the tubes Tn.

Still in Figure 3, it can be seen that the vapours from 31 are scrubbed in an annulus of liquid A and are absorbed by the carbamate solutions 32 being heated.

Through a thermosiphon effect the return circulation (arrows F) of the solution takes place in the central zone ZC of DC, separated from the bubbling zone A by a cylinder CI open at both ends.

In this central zone ZC, the carbamate solution is cooled in counter-current transferring the heat to the urea solution U in the distillation phase flowing through tubes Tn. In a second form of embodiment (Fig. 4), recovery steam is produced at 6 bar abs at least, as against 4.5 bar abs in existing plants. It has been found that it is possible to produce this steam in a quantity and quality sufficient for mean pressure distillation, by introducing a pre-condenser PRC (for example of the flowing hair or mane type) to which is sent at least 80% (from line 25) of the weak recycle carbamate solution, hence of the recycle water (line 24). With the high concentration of water in the carbamate solution so obtained, it is possible, at the same pressure, to raise the condensation heat to at least 170 °C, thus producing the desired steam at 6 bar at

east

In Figure 4 too the vacuum distiller-condenser D2 is the same as that in Figure 2. Preferably, an optimized version of this has been found, permitting the maximization of the heat transfer (Fig. 5).

It now consists of three sections: B1, B2 and B3. Said Figure 5 shows the whole of the apparatus which forms the vacuum distillation section (1st stage at 0.35 bar abs).

The upper part B1 operates with recovery steam VAR (from the carbamate condenser C*), whose condensate VC is discharged from the bottom of the above-mentioned zone B1, and a final concentration of 96% of urea in the distilled solution SU + V can thus be achieved.

The other part consists of two elements in series B2 and B3, in which the solution of carbamate (weak carbamate) 37 flows from top to bottom and is cooled in two distinct stages B2 and B3; in this way the final solution of carbamate 32 has the lowest possible temperature, hence minimum pressure.

The element B3 has besides an outlet for inert gas GI in its upper part.

The carbamate solution 37 as well as the residual vapour containing inert gas VR, are introduced, on the other hand, into ducts respectively 100 and 101 in the distiller-condenser in Figure 5.

Thanks to this pressure value (≈ 20 bar) the urea solution U can be distilled at mean pressure (20 bar) with recovery steam VAR at 6 bar in distiller MP.

The heat recoverable in the double effect system is not sufficient to distillate the solution up to 96% of urea. Consequently the distiller D2 is divided into two parts: an upper part B1 which uses recovery steam, and a lower part which in turn is subdivided into two zones B2 and B3, using the vapours containing inert gas VMP from the mean pressure distiller MP line 12 in Figure 4.

Operating conditions which can be achieved with the scheme described can be so summed up:

- Molar ratio NH_3/CO_2 in the reactor : 2.8
- Molar ratio $\text{H}_2\text{O}/\text{CO}_2$ in the reactor : 0.5
- Reactor yield : 57%
- Temperature at reactor outlet : 185°C
- Steam consumption at 20 bar : 800 kg/t
- Export recovery steam : 200 kg/t

In comparison to the reference case, a saving is achieved of 200 kg/t of steam at 20 bar, while there is a smaller recovery of 150 kg/t of steam at 4.5 bar.

With the addition of a mean pressure double-effect stage conditions can therefore be improved, even if not drastically.

In effect, the greatest obstacle to a significant improvement of the process lies in the low conversion yield of the reactor R, as a consequence of the low NH_3/CO_2 ratio in the reactor itself.

If the ratio is increased, this worsens operating conditions in the stripper ST, and therefore in the whole plant.

It has now been found that by adding a pre-distiller PRD upstream of the existing stripper ST the following results can be achieved:

- the possibility of feeding the stripper with a solution with a high NH_3/CO_2 ratio (> 2.5 weight), strippable with CO_2 ;
- the possibility, therefore, of operating with higher NH_3/CO_2 ratios in the reactor, thus increasing conversion yields;
- a substantial reduction in the stripper's heat load with CO_2 , with ample possibilities to overload.

Figure 6 shows the process scheme modified by the addition of a pre-distiller. As a preliminary step, an up-flow pre-distiller has been envisaged, so as avoid modification of the driving force of circulation of the solution.

The vapours 5 leaving the pre-distiller PRD are joined to those coming from the stripper ST, while the solution of partially distilled urea flows into the existing stripper for the final stripping with CO_2 .

Operating conditions figures have been calculated as follows:

- Molar ratio NH_3/CO_2 in the reactor : 3.4
- Molar ratio $\text{H}_2\text{O}/\text{CO}_2$ in the reactor : 0.5
- Reactor yield : 64%
- Temperature at reactor outlet : 185°C
- Reactor pressure : 141 bar
- Steam consumption at 20 bar : 820 kg/t
- Export steam at 4.5 bar : 120 kg/t.

The operating conditions obtained are therefore similar to those estimated for double effect.

According to a particularly advantageous aspect of the invention, the improvements achieved thanks to the adoption of the double-effect system (Figures 2 to 5) and to the addition of a pre-distiller (Figure 6) can

be combined, in the sense that their simultaneous application results substantially in the accumulation of the effects.

Figures 7 and 8 show the stripping process with CO₂ with the addition of a pre-distiller PRD and of the double-effect system, the latter in the two variations already described, i.e. in Figure 7 with the distiller-condenser DC from Figure 2, and in Figure 8 with the pre-condenser PRC from Figure 4.

The remarkable operating conditions which can be achieved are:

- Molar ratio NH₃/CO₂ in the reactor : 3.4
- Molar ratio H₂O/CO₂ in the reactor : 0.4
- Reactor yield : 64%
- Temperature at reactor outlet : 185 ° C
- Reactor pressure : 141 bar
- Steam consumption at 20 bar : 650 kg/t
- Export recovery steam at 4.5 bar : 100 kg/t.

By way of illustration but not of limitation the following balances or examples in Tables I, II and III are set out here.

EXAMPLE 1 (Table I) - Conventional process (Figure 1)

EXAMPLE 2 (Table II) - Double-effect process (Figures 2 and 4)

EXAMPLE 3 (Table III) - Process with pre-distiller (Figure 6)

CAPACITY - 1000 MTD UREA

 $\text{NH}_3/\text{CO}_2 = 2.8$
 $\text{H}_2\text{O}/\text{CO}_2 = 0.4$

REFERENCE CASE

TABLE 1

page 1

LINE	1		2		3		4		5		6	
Physical stage	Liquid		gas		liquid		vapours		liquid		vapours	
P (kg/cm ² abs.)	160		141		141		141		141		141	
T (°C)	40		100		183		180		170		160	
COMPOSITION	kg/h	zw	kg/h	zw	kg/h	zw	kg/h	zw	kg/h	zw	kg/h	zw
Urea												
NH ₃	23625	100,00			43403	34,62			41667	57,10		
CO ₂			30555	100,00	35814	28,56	31617	38,11	5181	7,10	1055	81,00
H ₂ O					24011	19,15	49133	59,22	6706	9,19	183	14,40
					22158	17,67	2219	2,67	19418	26,61	33	2,60
TOTAL	23625	100,00	30555	100,00	125386	100,00	82969	100,00	72972	100,00	1271	100,00

LINE	7		8		9		10		11		12	
Physical stage	vapours		liquid		vapours		liquid		liquid		vapours	
P (kg/cm ² abs.)	141				141		141		3,5		3,5	
T (°C)	183				167		167		135		135	
COMPOSITION	kg/h	zw	kg/h	zw	kg/h	zw	kg/h	zw	kg/h	zw	kg/h	zw
Urea												
NH ₃	5438	53,45	10619	36,64	13895	52,14	51966	47,71	41667	69,12	4388	34,59
CO ₂	4425	43,50	11131	38,41	11990	44,99	48274	44,32	313	0,52	6193	50,40
H ₂ O	310	3,05	7228	24,95	765	2,87	8682	7,97	17514	29,05	1904	15,01
TOTAL	10173	100,00	28978	100,00	26650	100,00	108922	100,00	60287	100,00	12685	100,00

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CAPACITY - 1000 MTD UREA

page 2

PRELIMINARY

TABLE 1

CO₂ STRIPPING PROCESS

REFERENCE CASE

NH₃/CO₂ = 2,8H₂O/CO₂ = 0,4

Z = 57 Z

LINE	13	14	15	16	17	18
Physical stage	Liquid	vapours	liquid	vapours	liquid	vapours
P(kg/cm ² abs.)	1,0	1,0	0,3	0,3	0,05	0,05
T (°C)	92	100	125	125	140	140
COMPOSITION	kg/h	kg/h	kg/h	kg/h	kg/h	kg/h
Urea	41667	73,08	41667	96,00	41667	99,8
NH ₃	336	0,99	457	13,97	336	2,47
CO ₂	114	0,20	199	6,08	114	0,84
H ₂ O	14899	26,13	2615	79,95	13163	96,69
TOTAL	57016	100,00	3271	100,00	41750	100,00

LINE	19	20	21	22	23
Physical stage	liquid	liquid	liquid		
P(kg/cm ² abs.)	3,5	3,5			
T (°C)	50	71			
COMPOSITION	kg/h	kg/h	kg/h	kg/h	kg/h
Urea	1848	25,00	6236	31,06	
NH ₃	496	6,71	6889	34,31	
CO ₂	5047	68,29	6951	34,63	
H ₂ O			12417	100,00	
TOTAL	7391	100,00	20076	100,00	

CAPACITY - 1000 MTD

DOUBLE EFFECT CASE

PRELIMINARY

page 1

NH₃/CO₂ = 2.8H₂O/CO₂ = 0.5

Z = 57 %

inerts = 0

losses = 0

TABLE 2

LINE	1		2		3		4		5		6	
Physical stage	Liquid		Gas		Liquid		vapours		Liquid		vapours	
P (kg/cm ² abs.)	160		141		141		141		141		141	
T (°C)	40		100		185		180		168		160	
COMPOSITION	kg/h	Zw	kg/h	Zw	kg/h	Zw	kg/h	Zw	kg/h	Zw	kg/h	Zw
Urea	23625		100,00		43403		34,00		41667		50,45	
NH ₃					35814		28,05		8672		10,50	
CO ₂			30555		24011		18,81		11149		13,50	
H ₂ O					24443		19,14		21100		25,55	
TOTAL	23625		100,00		127671		100,00		82588		100,00	

LINE	7		8		9		10		11		12	
Physical stage	vapours								Liquid		vapours	
P (kg/cm ² abs.)	141								19		19	
T (°C)	185								156		159	
COMPOSITION	kg/h	Zw	kg/h	Zw	kg/h	Zw	kg/h	Zw	kg/h	Zw	kg/h	Zw
Urea	5438								41667		58,95	
NH ₃	4425		53,45						5654		8,00	
CO ₂	310		43,50						3887		5,50	
H ₂ O			3,05						19470		25,50	
TOTAL	10173		100,00						70678		100,00	

CAPACITY - 1000 MTD

NH₃/CO₂ = 2,8H₂O/CO₂ = 0,5

Z = 57 %

PRELIMINARY

TABLE 2

DOUBLE EFFECT CASE

page 2

inerts = 0

losses = 0

LINE	13	14	15	16	17	18
Physical stage	liquid	vapours	liquid	vapours	liquid	vapours
P (kg/cm ² abs.)	3,5	3,5	1	1	0,3	0,3
T (°C)	135	135	92	92	125	125
COMPOSITION	kg/h	kg/h	kg/h	kg/h	kg/h	kg/h
Urea	41667	69,12	41667	73,08	41667	96,00
NH ₃	793	1,31	336	0,59	336	2,47
CO ₂	313	0,52	114	0,20	114	0,84
H ₂ O	17514	29,05	14899	26,13	1736	96,69
TOTAL	60287	100,00	57016	100,00	43403	100,00

LINE	19	20	21	22	23	24
Physical stage	liquid	vapours	liquid	liquid	liquid	liquid
P (kg/cm ² abs.)	0,05	0,05		3,5	3,5	141
T (°C)	140	140		50	40	120
COMPOSITION	kg/h	kg/h	kg/h	kg/h	kg/h	kg/h
Urea	41667	99,8		793	5654	9727
NH ₃				313	3887	11332
CO ₂	83	0,2		5014	6970	8633
H ₂ O		1653	12417	81,93	42,22	29,07
TOTAL	41750	100,00	12417	100,00	16511	29692

CAPACITY - 1000 MTD

$$\text{NH}_3/\text{CO}_2 = 2,8$$
$$\text{H}_2\text{O}/\text{CO}_2 = 0,5$$

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PRELIMINARY
inerts = 0
losses = 0

TABLE 2

DOUBLE EFFECT CASE

LINE	25	26								
Physical stage	Liquid	Liquid								
P (kg/cm ² abs.)	141	141								
T (°C)	120	120								
COMPOSITION	kg/h	Σw	kg/h	Σw	kg/h	Σw	kg/h	Σw	kg/h	Σw
Urea										
NH ₃	8249	32,76	1478	32,75						
CO ₂	9611	38,17	1721	38,13						
H ₂ O	7319	29,07	1314	29,12						
TOTAL	25179	100,00	4513	100,00						

[illegible]

LINE	1		2		3		4		5		6	
	kg/h	%w	kg/h	%w	kg/h	%w	kg/h	%w	kg/h	%w	kg/h	%w
Physical stage												
P (kg/cm ² abs.)												
T (°C)												
COMPOSITION												
Urea												
NH ₃												
CO ₂												
H ₂ O												
TOTAL												

LINE	7	8	9	10	11	12
Physical stage	liquid				vapours	vapours
P (kg/cm ² abs.)	141				141	141
T (°C)	170				185	160
COMPOSITION	kg/h	kg/h	kg/h	kg/h	kg/h	kg/h
	Zw	Zw	Zw	Zw	Zw	Zw
Urea	41667	55,00			5438	83,00
NH ₃	6060	8,00			43,50	14,40
CO ₂	7576	10,00			310	2,60
H ₂ O	20454	27,00			10173	100,00
TOTAL	75757	100,00			10173	100,00

CAPACITY - 1000 MTD UREA

$\text{NH}_3/\text{CO}_2 = 3,4$
 CASE WITH PREDISTILLER
 AND STRIPPER
 $\text{H}_2\text{O}/\text{CO}_2 = 0,5$
 $\Sigma = 64 \text{ t}$

TABLE 3

PRELIMINARY

PAGE 2

inerts = 0
 losses = 0

LINE	13	14	15	16	17	18
Physical stage	Liquid	vapours	liquid	vapours	liquid	vapours
P (kg/cm ² abs.)	3,5	3,5	1,0	1	0,3	0,3
T (°C)	135	135	92	100	125	125
COMPOSITION	kg/h	kg/h	kg/h	kg/h	kg/h	kg/h
Urea	41667	69,12	41667	73,08	41667	96,00
NH ₃	793	1,31	336	0,59	457	13,17
CO ₂	313	0,52	114	0,20	199	6,08
H ₂ O	17514	29,05	14899	26,13	2615	79,95
TOTAL	60287	100,00	57016	100,00	43403	100,00

LINE	19	20	21	22	23	24
Physical stage	Liquid	vapours	liquid	liquid	liquid	
P (kg/cm ² abs.)	0,05	0,05	-	3,5	141	
T (°C)	140	140	-	50	71	
COMPOSITION	kg/h	kg/h	kg/h	kg/h	kg/h	kg/h
Urea	41667	99,8		1848	7115	31,12
NH ₃				496	7759	33,94
CO ₂				5047	7987	34,94
H ₂ O	83	1653	12417	68,29	100,00	
TOTAL	41750	100,00	12417	100,00	22861	100,00

Claims

1. Process for the production of urea comprising at least: a stage wherein ammonia and carbon dioxide are reacted in a reactor at a pressure of between 120 and 220 bar and temperature of between 170 °C and 200 °C, a stripping stage wherein the urea solution leaving the reactor and containing carbamate and water and other products, reacted or unreacted, undergoes a process to strip it of carbamate and NH₃ by means of the carbon dioxide feeding the reactor; a stage wherein the stripped vapours are condensed and two vacuum distillation stages, characterized by the fact that it comprises at least one additional stage selected from the group consisting of: a mean pressure distillation stage using in substance recovery heat, by preference of the double-effect type, a pre-distillation stage and a second stripping stage.
2. Process according to claim 1, characterized by the fact that the distillation heat is produced by direct condensation of the vapours forming carbamate in a condenser-distiller wherein vapour condensation takes place on the shell side, and the heat produced is used to distill the urea solution flowing inside the tubes.
3. Process according to claim 2, characterized by the fact that the vapours bubble in an annulus of liquid and are absorbed by the carbamate solution which is thereby heated, and by thermosiphon effect the solution flows by return circulation into a central zone where the carbamate solution is cooled in counter-current transferring heat to the solution.
4. Process according to claim 1, characterized by the fact that recovery vapour is produced at 6 bar abs at least in a quantity and of a quality sufficient for distillation.
5. Process according to claim 4, characterized by the fact that a pre-condenser is used, into which is directed at least 80% of the carbamate solution, so that the condensation heat can be raised to at least 170 °C thus making it possible to produce vapour at 6 bar abs at least.
6. Process according to claims 2 and 5, characterized by the fact that a distiller-condenser in three superimposed sections is used, in which the urea solution is concentrated on the tube side up to 92-97% weight at the upper section outlet, whereas on the shell side the heat is provided in the upper section by condensing the recovery vapour, and in the intermediate and lower sections by condensing vapours to form carbamate in two distinct sections in counter-current to the urea solution so that the condensation of the above vapours is effected at the minimum possible temperature and pressure.
7. Process according to claim 1, characterized by the fact that a pre-distiller is used.
8. Process according to claim 1, characterized by the combination of a pre-distiller and of a double-effect distiller-condenser.

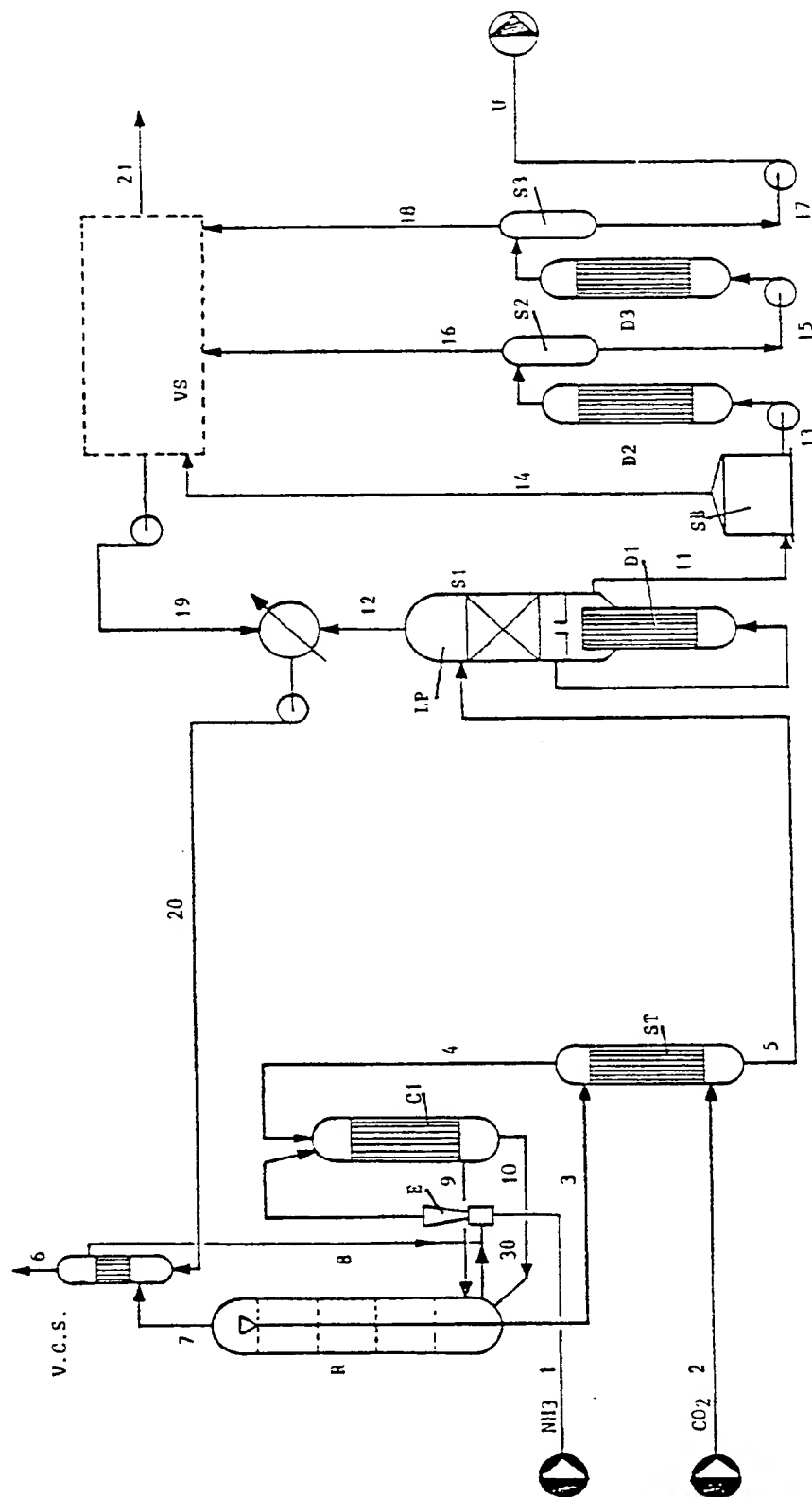


Fig. 1

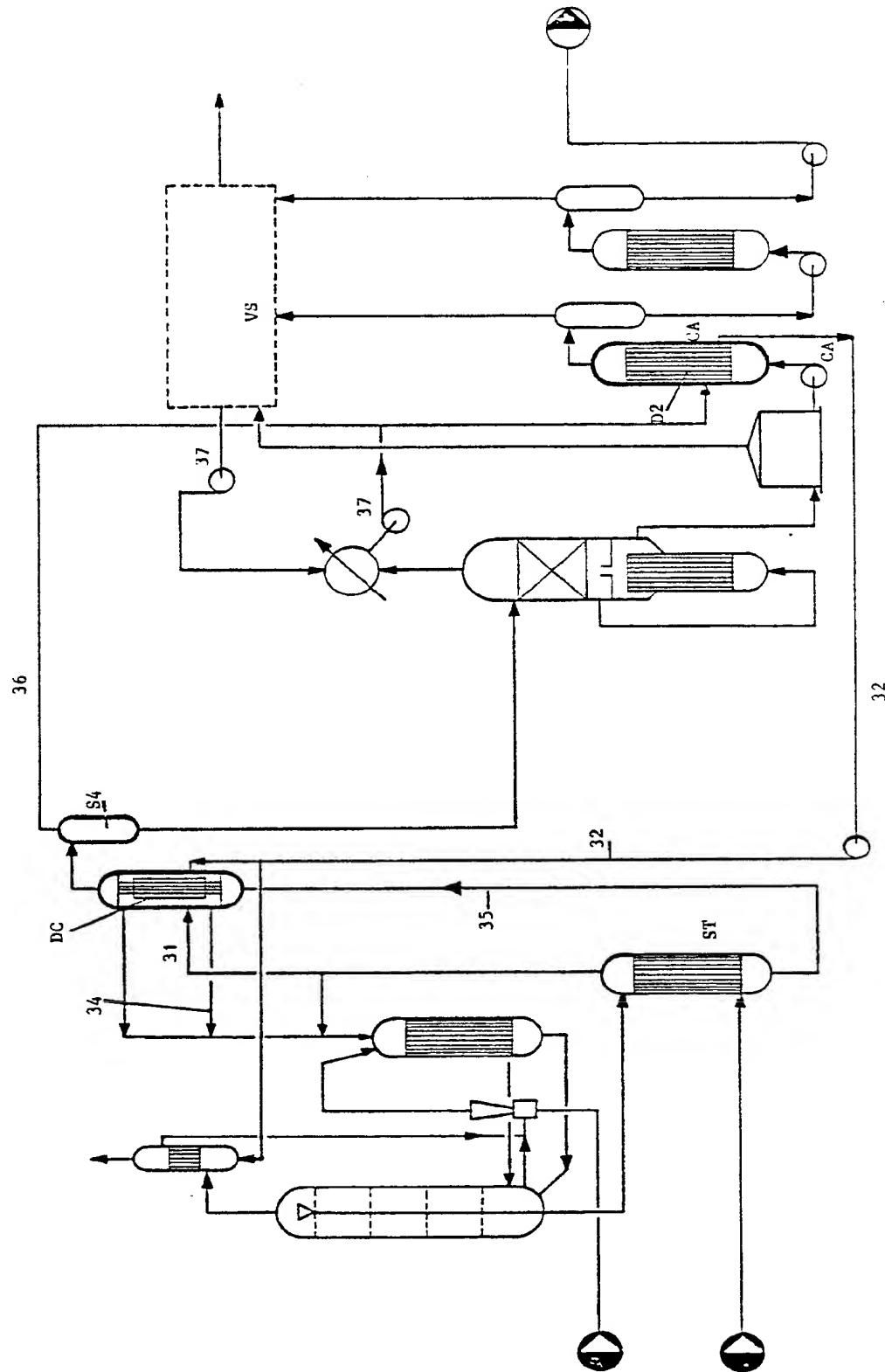


Fig. 2

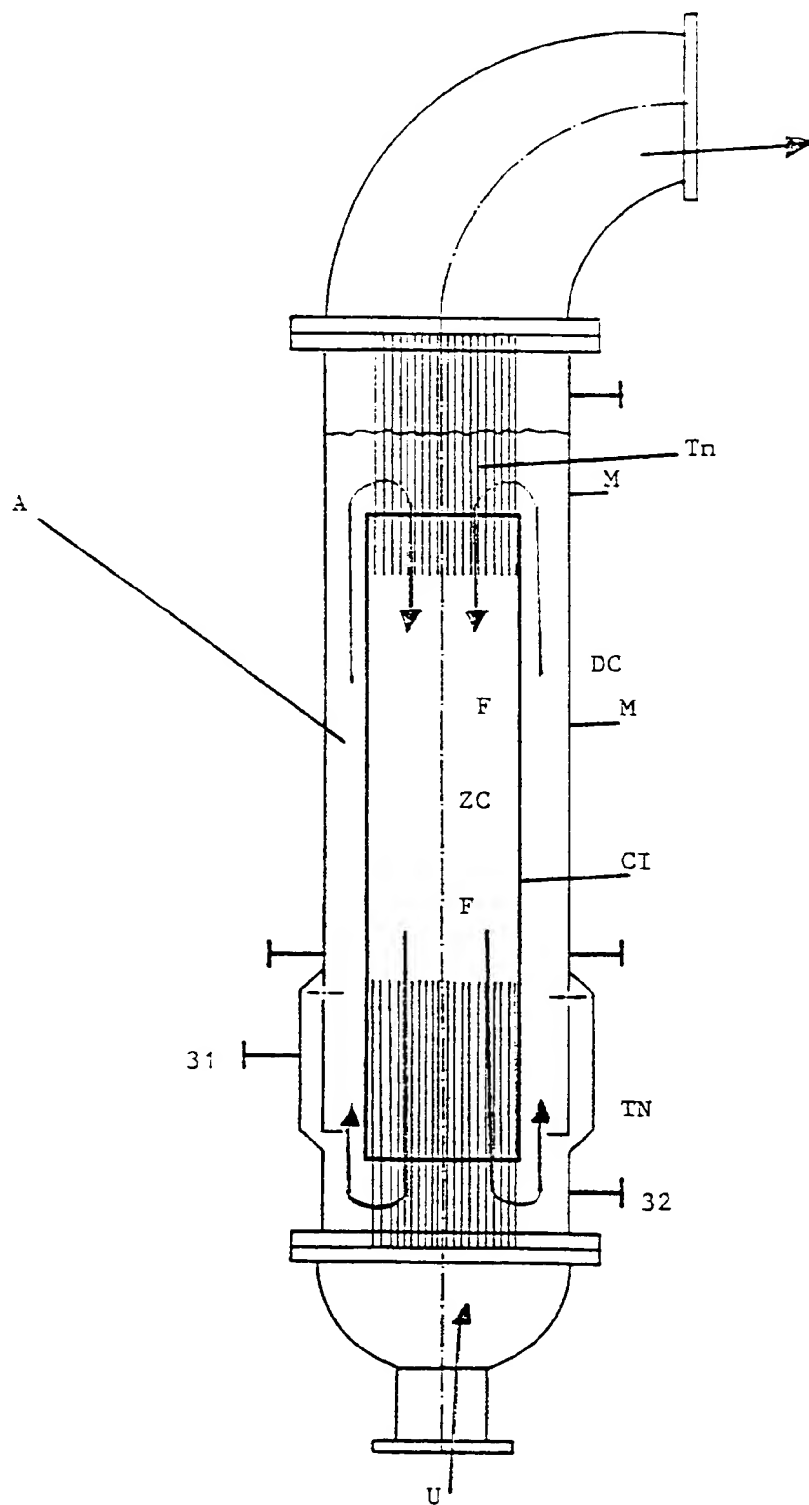


Fig. 3

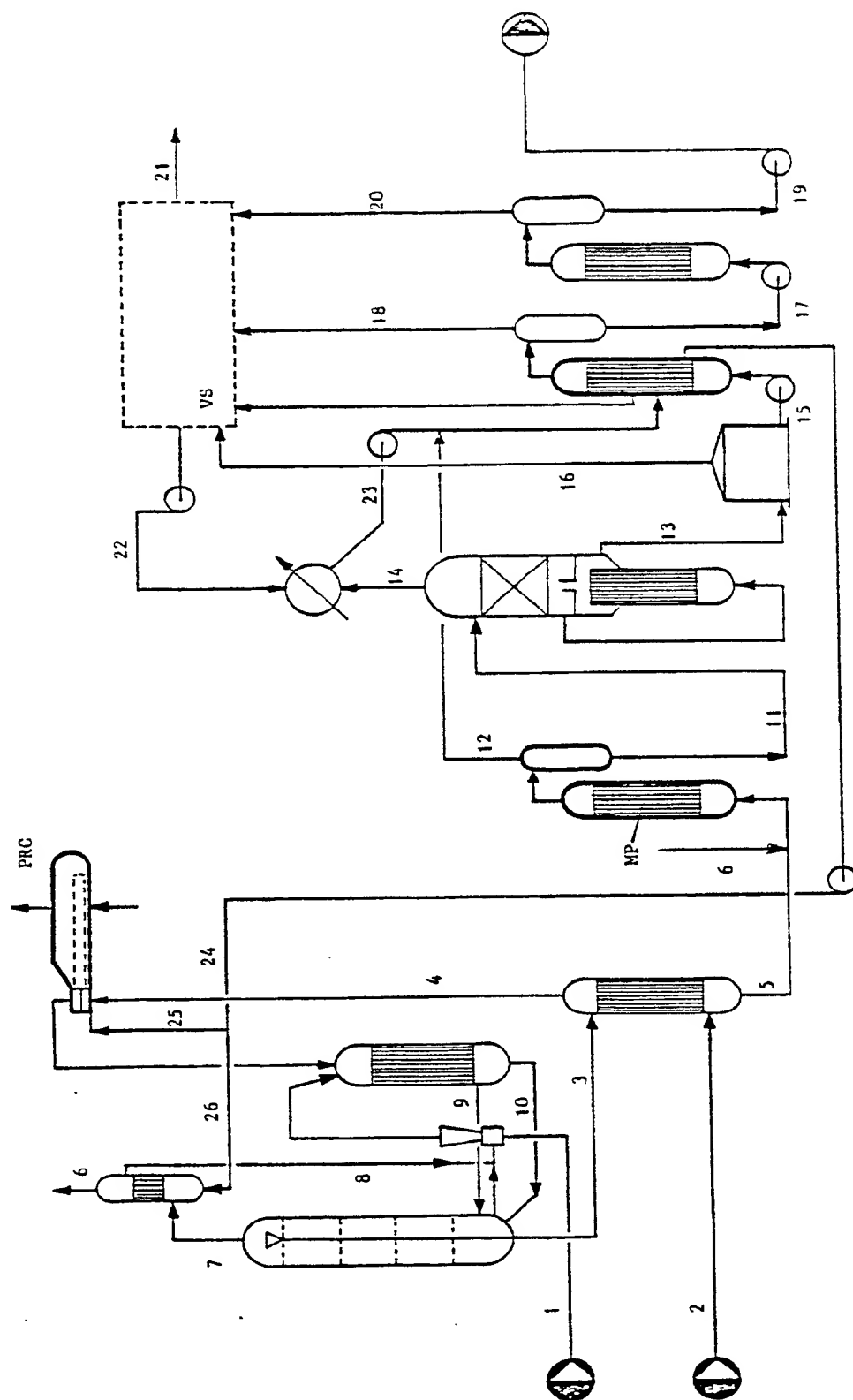


Fig. 4

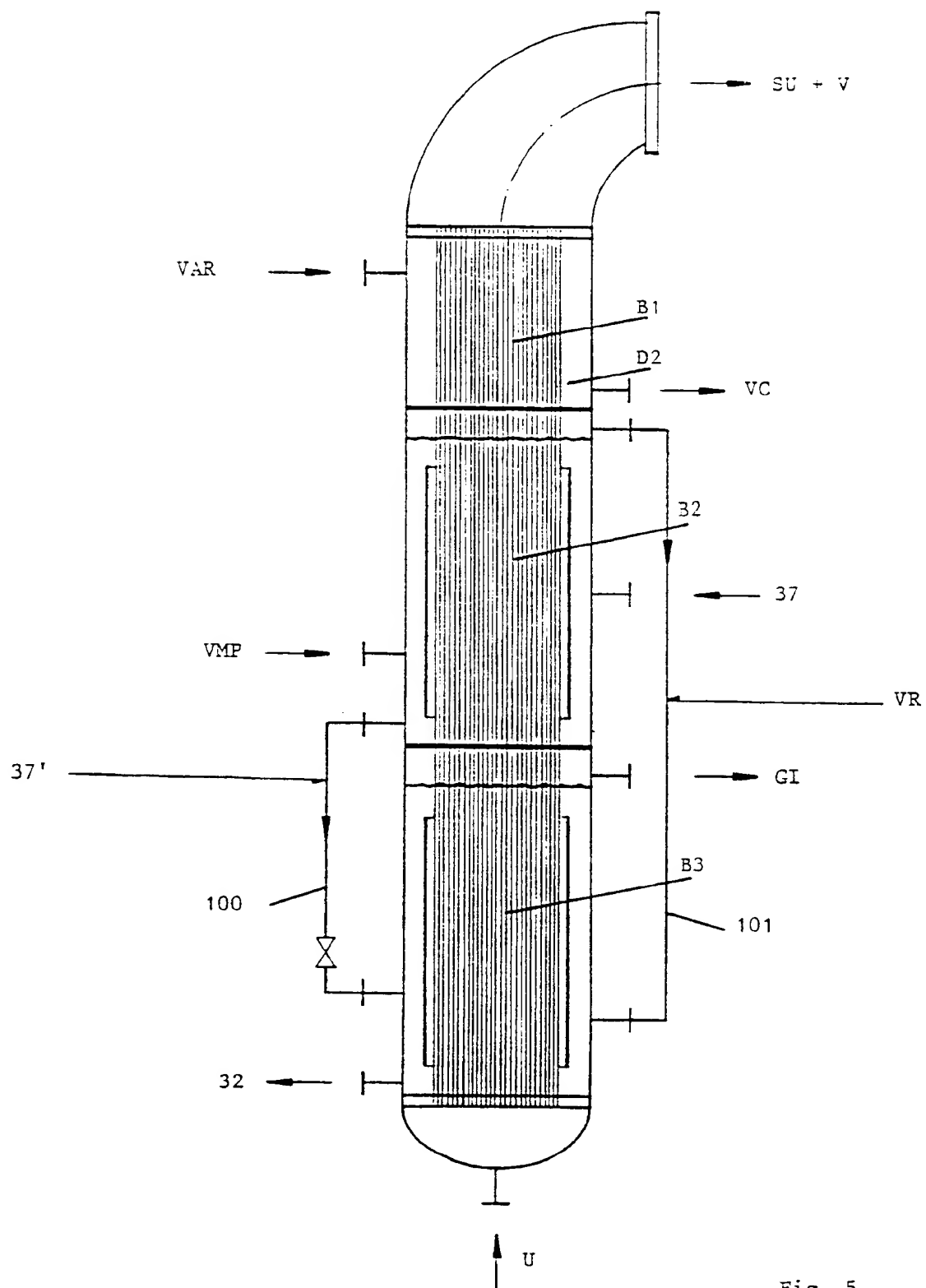


Fig. 5

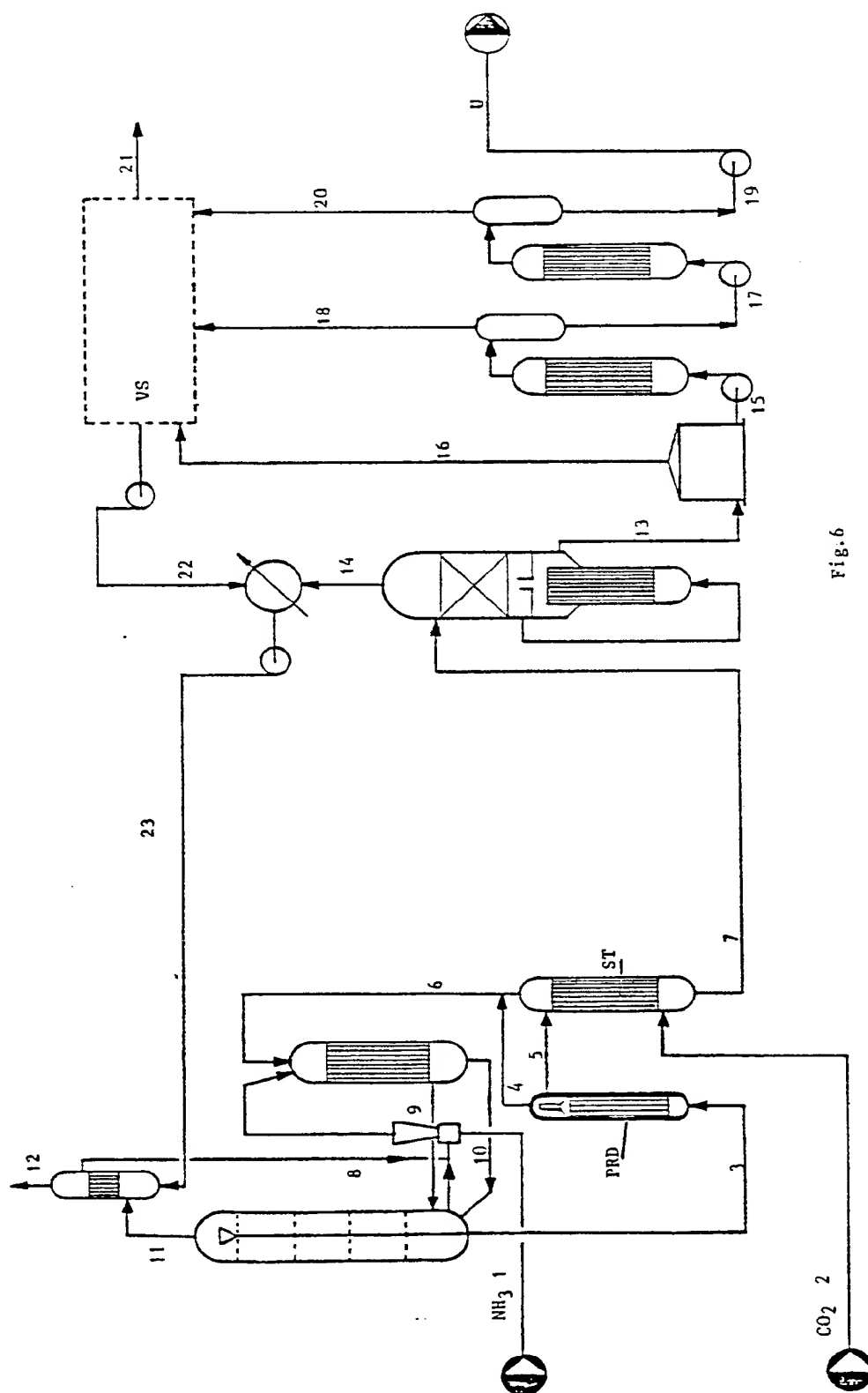


Fig. 6

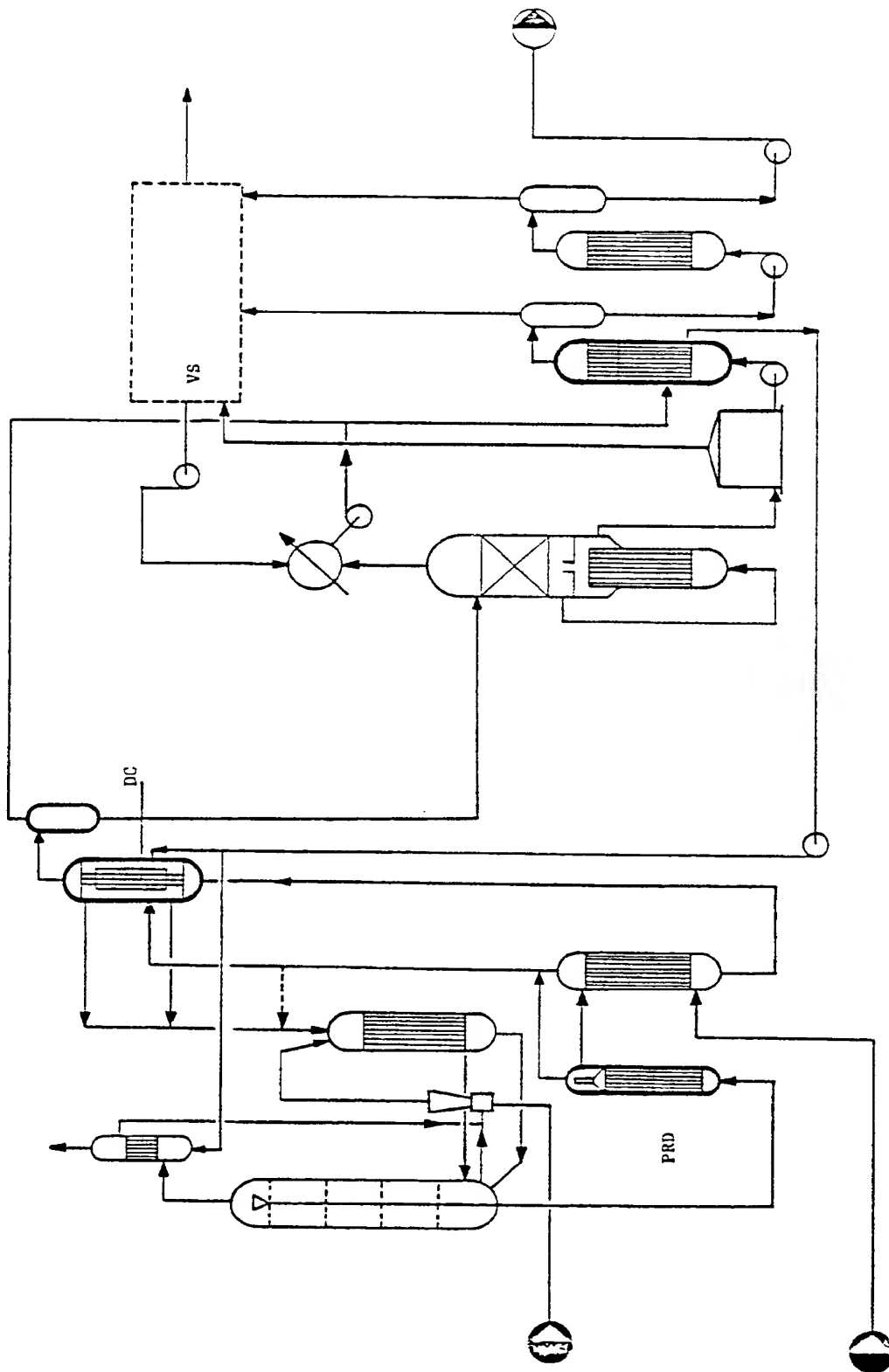


Fig. 7

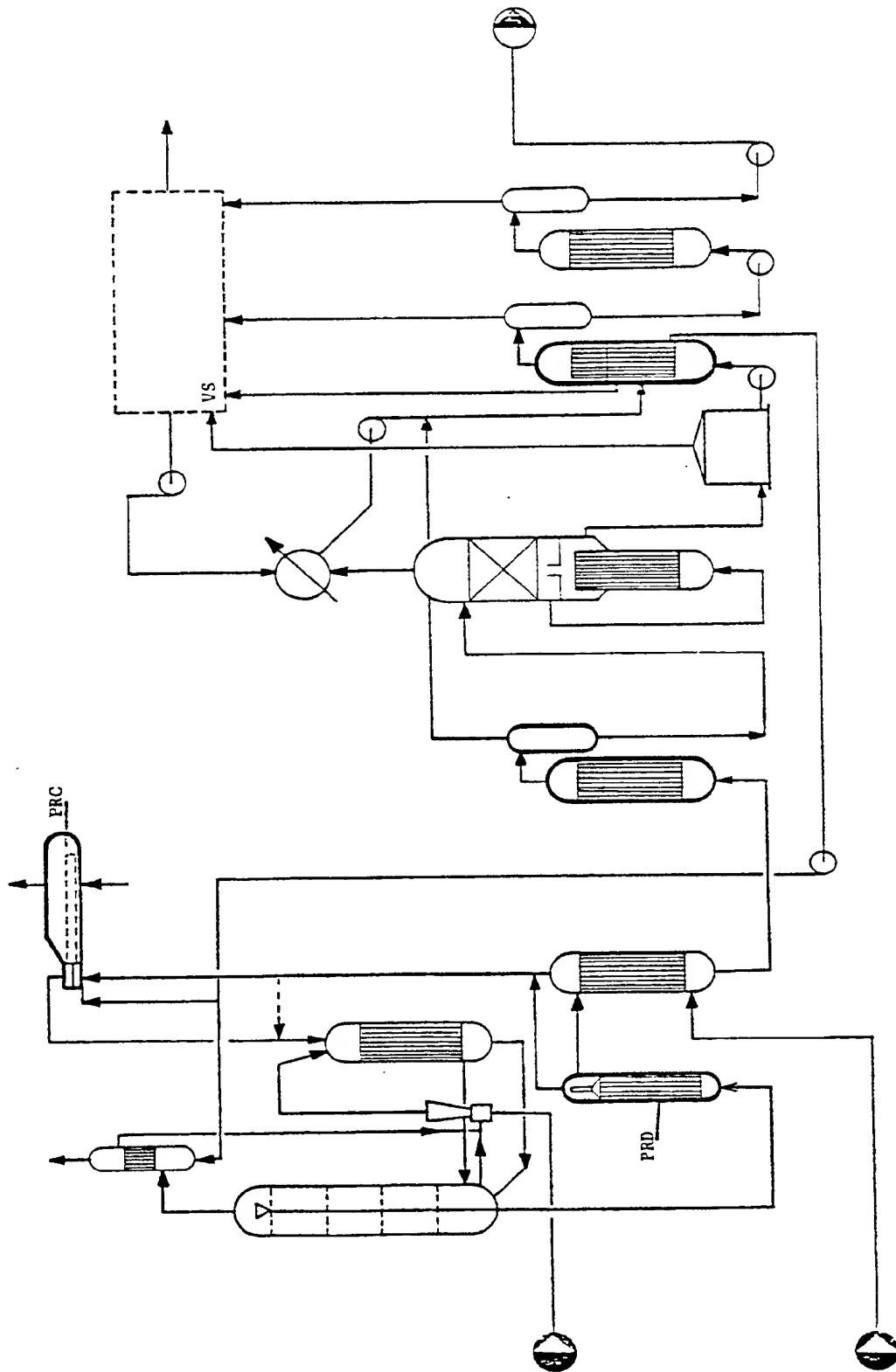
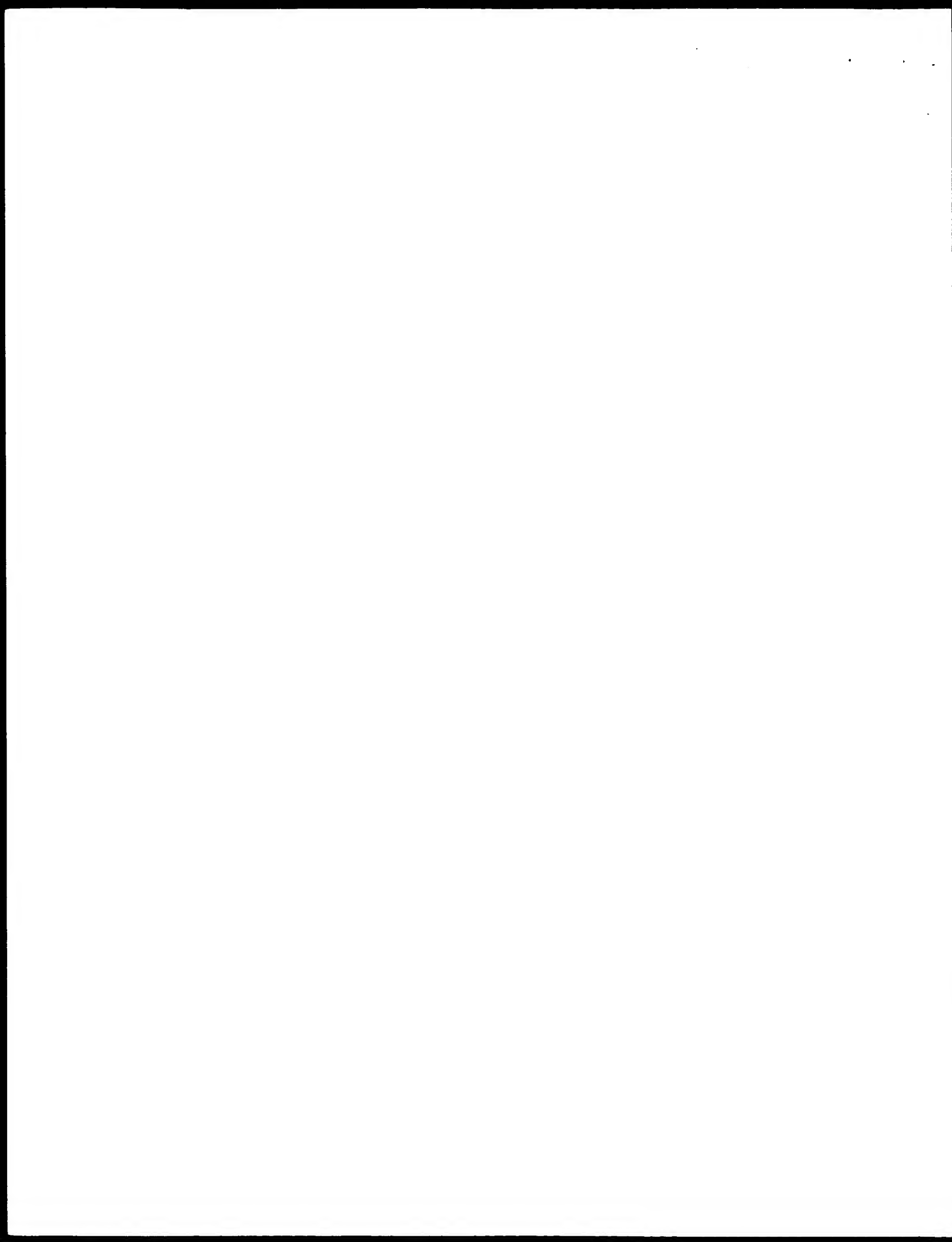


Fig. 8



(19)



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15.11.89 Bulletin 89/46(71) Applicant: **AMMONIA CASALE S.A.**
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AMMONIA CASALE S.A. Via della Posta 4
CH-6900 Lugano(CH)(54) **Improvements of processes and plants for the production of urea.**

(57) To the conventional process for the production of urea, comprising a stage for the reaction of NH_3 and CO_2 , a stage for stripping with CO_2 the urea solution containing carbamate and other products, a stage for condensing the stripped vapours and two stages of vacuum distillation, is now added at least a further stage selected from the group consisting of: a mean pressure distillation stage with substantially recovery heat, more particularly with double effect, a pre-distillation stage and a second stripping stage.

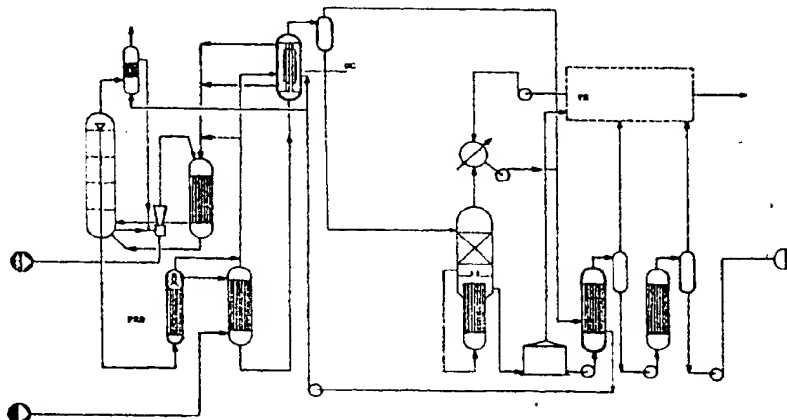


Fig. 1

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EUROPEAN SEARCH REPORT

Application Number

EP 88 10 7081

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 4)
A	US-A-4 256 662 (GORLOVSKY et al.) ---		C 07 C 126/02
A	US-A-3 824 283 (SHUNICHI HARADA et al.) ---		
A	DE-A-1 468 245 (MONTECATINI) -----		
			TECHNICAL FIELDS SEARCHED (Int. Cl. 4)
			C 07 C 126/00
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 05-09-1989	Examiner VAN GEYT J.J.A.
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	